

# Influence of lattice structure on multipole interactions in $\Gamma_3$ non-Kramers doublet systems

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We study the multipole interactions between  $f^2$  ions with the  $\Gamma_3$  non-Kramers doublet ground state under a cubic crystalline electric field. We construct the  $\Gamma_3$  doublet state of the electrons with the total angular momentum  $j = 5/2$ . By applying the second-order perturbation theory with respect to the intersite hopping, we derive the multipole interactions. We obtain a quadrupole interaction for a simple cubic lattice, an octupole interaction for a bcc lattice, and both quadrupole and octupole interactions for an fcc lattice. We also discuss general tendencies of the multipole interactions depending on the lattice structure by comparing the results with those for the  $\Gamma_8$  quartet systems of  $f^1$  ions.

## I. INTRODUCTION

In the field of the  $f$ -electron systems, the phenomena which originate from the multipole degrees of freedom have been studied intensively since such degrees of freedom, in addition to the dipole, are expected to become sources of exotic ordering and physical properties. The quadrupole moment couples to the lattice and its influence can be detected, for example, by ultrasonic measurements. In recent years, even the effects of the octupole moment have been investigated.

One of the most representative phenomena discovered in multipole physics is the quadrupole and octupole ordering in  $\text{NpO}_2$ <sup>1-5</sup> and  $\text{Ce}_x\text{La}_{1-x}\text{B}_6$ .<sup>6-12</sup> While it is in general difficult to detect the octupole moment, resonant x-ray scattering,<sup>1,10</sup> NMR,<sup>2</sup> anisotropic magnetization,<sup>9</sup> and neutron scattering<sup>11</sup> experiments have confirmed the octupole order. In these compounds, the crystalline electric field (CEF) ground state is the  $\Gamma_8$  quartet, which has sufficient degrees of freedom to possess quadrupole and octupole moments in addition to the dipole moment. Then, the  $\Gamma_8$  quartet has been regarded as an ideal system for multipole physics.

However, large degeneracy, such as in a quartet, is not a necessary condition to possess higher-order multipole moments. If the CEF ground state does not have the dipole moment but is not a singlet, this state inevitably has the higher-order multipole degrees of freedom. In fact, the  $\Gamma_3$  doublet state under a cubic CEF, which we will explore in this paper, does not have the dipole but has the quadrupole moments  $O_2^0$  and  $O_2^2$  with the  $\Gamma_{3g}$  symmetry and the octupole moment  $T_{xyz}$  with the  $\Gamma_{2u}$  symmetry. The absence of the dipole moment is also an advantage of the  $\Gamma_3$  systems since we can focus only on the higher-order multipoles. In the  $\Gamma_3$  state, the degeneracy is not due to the Kramers theorem, which is applicable only to an ion with odd number of  $f$  electrons. Thus, we consider an ion with even number of  $f$  electrons.

In particular,  $\text{Pr}^{3+}$  ion has two  $f$  electrons and in some Pr compounds, the CEF ground state is the  $\Gamma_3$  doublet. In recent years, interesting phenomena, which

probably originate from the multipole degrees of freedom, have been reported for Pr compounds with the  $\Gamma_3$  CEF ground state. In  $\text{PrPb}_3$ , incommensurate quadrupole ordering has been reported.<sup>13</sup>  $\text{PrIr}_2\text{Zn}_{20}$  and some of other Pr compounds with the same crystal structure (Pr 1-2-20 compounds) with the  $\Gamma_3$  CEF ground state become superconducting at low temperatures,<sup>14-19</sup> which might be mediated by multipole fluctuations.

In this paper, to elucidate multipole phenomena of the  $\Gamma_3$  systems, we derive the multipole interactions from a simple model only with  $f$ - $f$  direct hopping. While the actual exchange process would be through orbitals other than the  $f$  orbital, such a process can be represented by effective  $f$ - $f$  hopping. An important point is that the symmetry of the  $f$  orbital restricts the form of the hopping for both cases of the direct and effective hopping and we will obtain qualitatively the same results for the multipole interactions.<sup>5</sup>

The anisotropy in the multipole moments are closely tied to the real space direction and the multipole interactions are intrinsically anisotropic. It is in sharp contrast to the isotropic spin-spin interaction in a system without spin-orbit coupling. Thus, the nature of the multipole interactions can depend drastically on lattice structure. In the present study, we pay attention to this point of the multipole interactions. Then, we derive the multipole interactions for simple cubic (sc), bcc, and fcc lattices. We also compare the results of the present model for the  $f^2$ - $\Gamma_3$  systems with those of the  $\Gamma_8$  model for the  $f^1$  systems<sup>3,4</sup> to find common features among these two classes.

## II. GROUND AND INTERMEDIATE STATES

To construct electronic states, we first include the effect of the spin-orbit coupling in the one-electron states and consider only the  $f$ -electron states with the total angular momentum  $j = 5/2$ . These states split into the states with  $\Gamma_7$  and  $\Gamma_8$  symmetry under a cubic CEF [see Fig. 1(a)]. The  $\Gamma_7$  states at site  $\mathbf{r}$  are given by

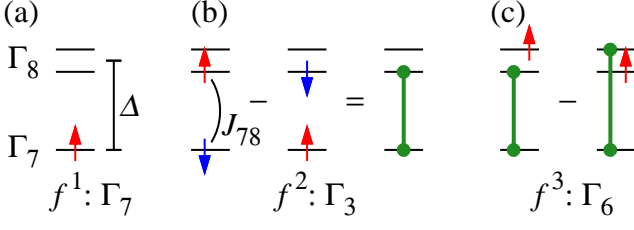


FIG. 1. (Color online) Electron configurations for (a)  $\Gamma_7$  state of  $f^1$ , (b)  $\Gamma_3$  state of  $f^2$ , and (c)  $\Gamma_6$  state of  $f^3$ . The bold lines denote spin singlets composed of the  $\Gamma_7$  and  $\Gamma_8$  orbitals.

$$c_{\mathbf{r}\uparrow}^\dagger|0\rangle \equiv \frac{1}{\sqrt{6}} \left( a_{\mathbf{r}5/2}^\dagger - \sqrt{5}a_{\mathbf{r}-3/2}^\dagger \right) |0\rangle, \quad (1a)$$

$$c_{\mathbf{r}\downarrow}^\dagger|0\rangle \equiv \frac{1}{\sqrt{6}} \left( a_{\mathbf{r}-5/2}^\dagger - \sqrt{5}a_{\mathbf{r}3/2}^\dagger \right) |0\rangle, \quad (1b)$$

where  $a_{\mathbf{r}j_z}^\dagger$  is the creation operator of the electron with the  $z$ -component  $j_z$  of the total momentum at  $\mathbf{r}$  and  $|0\rangle$  denotes the vacuum state. The  $\Gamma_8$  states are given by

$$c_{\mathbf{r}\alpha\uparrow}^\dagger|0\rangle \equiv \frac{1}{\sqrt{6}} \left( \sqrt{5}a_{\mathbf{r}5/2}^\dagger + a_{\mathbf{r}-3/2}^\dagger \right) |0\rangle, \quad (2a)$$

$$c_{\mathbf{r}\alpha\downarrow}^\dagger|0\rangle \equiv \frac{1}{\sqrt{6}} \left( \sqrt{5}a_{\mathbf{r}-5/2}^\dagger + a_{\mathbf{r}3/2}^\dagger \right) |0\rangle, \quad (2b)$$

$$c_{\mathbf{r}\beta\uparrow}^\dagger|0\rangle \equiv a_{\mathbf{r}1/2}^\dagger|0\rangle, \quad (2c)$$

$$c_{\mathbf{r}\beta\downarrow}^\dagger|0\rangle \equiv a_{\mathbf{r}-1/2}^\dagger|0\rangle. \quad (2d)$$

In the above equations,  $\sigma = \uparrow$  or  $\downarrow$  denotes the Kramers degeneracy of the one-electron states, while it is not a real spin because of the spin-orbit coupling. In the following, however, we call it spin for simplicity.

In actual situations, the  $f^2$ - $\Gamma_3$  doublet is mainly composed of two singlets between  $\Gamma_7$  and  $\Gamma_8$  orbitals, and then, we assume an antiferromagnetic interaction between the  $\Gamma_7$  and  $\Gamma_8$  orbitals. Such an interaction would be justified by perturbatively including the effects of the sixth order terms in the CEF, which cannot be included as a one-electron potential for  $j = 5/2$  states but are indispensable to stabilize the  $\Gamma_3$  state.<sup>20</sup>

The model Hamiltonian is

$$H = H_{\text{kin}} + H_{\text{loc}}. \quad (3)$$

$H_{\text{kin}}$  is the kinetic energy term which we will discuss later. The local part is given by

$$H_{\text{loc}} = \Delta \sum_{\mathbf{r}} (n_{\mathbf{r}8} - n_{\mathbf{r}7}) + J_{78} \sum_{\mathbf{r}} \mathbf{s}_{\mathbf{r}7} \cdot \mathbf{s}_{\mathbf{r}8}, \quad (4)$$

where

$$n_{\mathbf{r}7} = \sum_{\sigma} c_{\mathbf{r}7\sigma}^\dagger c_{\mathbf{r}7\sigma}, \quad (5a)$$

$$n_{\mathbf{r}8} = \sum_{\tau\sigma} c_{\mathbf{r}8\tau\sigma}^\dagger c_{\mathbf{r}8\tau\sigma}, \quad (5b)$$

$$\mathbf{s}_{\mathbf{r}7} = \frac{1}{2} \sum_{\sigma\sigma'} c_{\mathbf{r}7\sigma}^\dagger \boldsymbol{\sigma}_{\sigma\sigma'} c_{\mathbf{r}7\sigma'}, \quad (5c)$$

$$\mathbf{s}_{\mathbf{r}8} = \frac{1}{2} \sum_{\tau\sigma\sigma'} c_{\mathbf{r}8\tau\sigma}^\dagger \boldsymbol{\sigma}_{\sigma\sigma'} c_{\mathbf{r}8\tau\sigma'}. \quad (5d)$$

Here,  $\tau = \alpha$  or  $\beta$  and  $\boldsymbol{\sigma}$  are the Pauli matrices.  $\Delta$  denotes the CEF level splitting [see Fig. 1(a)] and  $J_{78}$  denotes the coupling constant of the antiferromagnetic interaction between the  $\Gamma_7$  and  $\Gamma_8$  orbitals [see Fig. 1(b)].

Then, for a sufficiently large  $J_{78}$ , the  $f^2$  ground states are spin singlets composed of the  $\Gamma_7$  and  $\Gamma_8$  orbitals [see Fig. 1(b)]:

$$\begin{aligned} |\tau(\mathbf{r})\rangle &\equiv \frac{1}{\sqrt{2}} (c_{\mathbf{r}\tau\uparrow}^\dagger c_{\mathbf{r}7\downarrow}^\dagger - c_{\mathbf{r}\tau\downarrow}^\dagger c_{\mathbf{r}7\uparrow}^\dagger) |0\rangle \\ &= \frac{i}{\sqrt{2}} \sigma_{\sigma\sigma'}^y c_{\mathbf{r}\tau\sigma}^\dagger c_{\mathbf{r}7\sigma'}^\dagger |0\rangle \\ &\equiv B_{\sigma\sigma'} c_{\mathbf{r}\tau\sigma}^\dagger c_{\mathbf{r}7\sigma'}^\dagger |0\rangle. \end{aligned} \quad (6)$$

The repeated indices should be summed hereafter. These states constitute a basis of the  $\Gamma_3$  representation of cubic symmetry.

Note that the present model is one of the simplest models to realize the  $\Gamma_3$  ground state and we should improve it if we deal with the CEF excited states. For example, when we accommodate two electrons in the  $\Gamma_8$  orbitals, we obtain six states with energy  $2\Delta$ , but they should split into three levels. To describe such splitting in the CEF excited states, it is necessary to include the interactions between  $\Gamma_8$  orbitals. Thus, we should restrict ourselves to low energy states around the  $\Gamma_3$  CEF ground state in the present simplified model.

We consider the exchange process between nearest-neighbor sites with the  $\Gamma_3$  ground state. Among the intermediate  $f^1$ - $f^3$  states, we consider only the lowest energy states. If the  $f^3$  site has zero or two  $\Gamma_7$  electrons, it cannot gain the energy from the antiferromagnetic interaction. Concerning the  $f^1$  states, we assume that the  $\Gamma_7$  state has lower energy, i.e.,  $\Delta > 0$ . However,  $\Delta$  should be sufficiently smaller than  $J_{78}$  for the realization of the  $\Gamma_3$  ground state in the  $f^2$  configurations. Then, each site should have one  $\Gamma_7$  electron in the intermediate states. That is, only the hopping between the  $\Gamma_8$  orbitals is allowed. In the following, we explicitly write the intermediate states and evaluate the matrix elements of the exchange processes.

The intermediate  $f^1$  states are the  $\Gamma_7$  states [see Fig. 1(a)],

$$|\sigma(\mathbf{r})\rangle \equiv c_{\mathbf{r}7\sigma}^\dagger |0\rangle. \quad (7)$$

We calculate the matrix elements of the annihilation operator of the  $\Gamma_8$  electron between the  $f^1$  and the  $\Gamma_3$

states. The effect of the annihilation operator on the  $\Gamma_3$  state is written as

$$\begin{aligned} c_{\mathbf{r}\tau\sigma}|\tau'(\mathbf{r})\rangle &= c_{\mathbf{r}\tau\sigma}B_{\sigma'\sigma''}c_{\mathbf{r}\tau'\sigma'}^\dagger c_{\mathbf{r}\tau'\sigma''}^\dagger|0\rangle \\ &= \delta_{\tau\tau'}B_{\sigma\sigma'}c_{\mathbf{r}\tau'\sigma'}^\dagger|0\rangle \\ &\equiv B_{\tau\sigma;\sigma'}^{\tau'}|\sigma'(\mathbf{r})\rangle. \end{aligned} \quad (8)$$

Then, we obtain the matrix element as

$$\langle\sigma'(\mathbf{r})|c_{\mathbf{r}\tau\sigma}|\tau'(\mathbf{r})\rangle = B_{\tau\sigma;\sigma'}^{\tau'}. \quad (9)$$

Note that  $B_{\tau\sigma;\sigma'}^{\tau'*} = B_{\tau\sigma;\sigma'}^{\tau'}$ , since we have defined these states with real coefficients from the basis  $\Gamma_7$  and  $\Gamma_8$  states.

The ground states among the  $f^3$  states for a strong antiferromagnetic interaction  $J_{78}$  between the  $\Gamma_7$  and  $\Gamma_8$  orbitals are the  $\Gamma_6$  states [see Fig. 1(c)],

$$\begin{aligned} |\uparrow(\mathbf{r})\rangle &\equiv \frac{1}{\sqrt{6}}(2c_{\mathbf{r}\alpha\uparrow}^\dagger c_{\mathbf{r}\beta\uparrow}^\dagger c_{\mathbf{r}\gamma\downarrow}^\dagger \\ &\quad - c_{\mathbf{r}\alpha\uparrow}^\dagger c_{\mathbf{r}\beta\downarrow}^\dagger c_{\mathbf{r}\gamma\uparrow}^\dagger - c_{\mathbf{r}\alpha\downarrow}^\dagger c_{\mathbf{r}\beta\uparrow}^\dagger c_{\mathbf{r}\gamma\uparrow}^\dagger)|0\rangle \end{aligned} \quad (10a)$$

$$\begin{aligned} &= \frac{1}{\sqrt{3}}[c_{\mathbf{r}\alpha\uparrow}^\dagger|\beta(\mathbf{r})\rangle - c_{\mathbf{r}\beta\uparrow}^\dagger|\alpha(\mathbf{r})\rangle], \\ |\downarrow(\mathbf{r})\rangle &\equiv \frac{1}{\sqrt{6}}(2c_{\mathbf{r}\alpha\downarrow}^\dagger c_{\mathbf{r}\beta\downarrow}^\dagger c_{\mathbf{r}\gamma\uparrow}^\dagger \\ &\quad - c_{\mathbf{r}\alpha\downarrow}^\dagger c_{\mathbf{r}\beta\uparrow}^\dagger c_{\mathbf{r}\gamma\downarrow}^\dagger - c_{\mathbf{r}\alpha\uparrow}^\dagger c_{\mathbf{r}\beta\downarrow}^\dagger c_{\mathbf{r}\gamma\downarrow}^\dagger)|0\rangle \end{aligned} \quad (10b)$$

$$= \frac{1}{\sqrt{3}}[-c_{\mathbf{r}\alpha\downarrow}^\dagger|\beta(\mathbf{r})\rangle + c_{\mathbf{r}\beta\downarrow}^\dagger|\alpha(\mathbf{r})\rangle].$$

Note that, in a local model considering all the 14  $f$ -orbitals, we obtain the  $\Gamma_6$  ground state when we accommodate three electrons for a realistic parameter set to obtain a  $\Gamma_3$  ground state in an  $f^2$  case.<sup>20</sup> Thus, the intermediate  $\Gamma_6$  state is reasonable. Note also that the states  $c_{\mathbf{r}\alpha\sigma}^\dagger|\beta(\mathbf{r})\rangle + c_{\mathbf{r}\beta\sigma}^\dagger|\alpha(\mathbf{r})\rangle$  are represented by (spin singlet composed of two  $\Gamma_8$  orbitals) $\otimes\Gamma_7$  and they do not gain the antiferromagnetic energy. The matrix element of the creation operator is given by

$$\langle\sigma'(\mathbf{r})|c_{\mathbf{r}\tau\sigma}^\dagger|\tau'(\mathbf{r})\rangle = i\sigma_{\tau\tau'}^y\sigma_{\sigma\sigma'}^z\frac{\sqrt{3}}{2} \equiv \tilde{B}_{\tau\sigma;\sigma'}^{\tau'}. \quad (11)$$

We note that  $\tilde{B}_{\tau\sigma;\sigma'}^{\tau'*} = \tilde{B}_{\tau\sigma;\sigma'}^{\tau'}$ .

### III. HOPPING

The hopping processes are described by the kinetic energy term of the Hamiltonian for the  $\Gamma_8$  orbitals:

$$\begin{aligned} H_{\text{kin}} &= \sum_{\mathbf{r}, \boldsymbol{\mu}, \tau, \sigma, \tau', \sigma'} c_{\mathbf{r}+\boldsymbol{\mu}\tau\sigma}^\dagger t_{\tau\sigma;\tau'\sigma'}^\boldsymbol{\mu} c_{\mathbf{r}\tau'\sigma'}, \\ &= \sum_{\mathbf{r}, \boldsymbol{\mu}, \nu, \nu'} c_{\mathbf{r}+\boldsymbol{\mu}\nu}^\dagger t_{\nu\nu'}^\boldsymbol{\mu} c_{\mathbf{r}\nu'}, \end{aligned} \quad (12)$$

where the vector  $\boldsymbol{\mu}$  connects nearest-neighbor sites. Here, we have introduced an abbreviation  $\nu = (\tau, \sigma)$ . Since  $H_{\text{kin}}$  is Hermitian,  $t_{\nu\nu'}^{\boldsymbol{\mu}*} = t_{\nu'\nu}^\boldsymbol{\mu}$ .

In this study, we consider only the  $\sigma$  bonding ( $ff\sigma$ ) for the hopping integrals. Although the hopping integrals were derived in Ref. 21 for the sc lattice and in Ref. 4 for the other lattices, here we write down again the hopping integrals for readers' convenience. To write out the hopping integral  $t^\boldsymbol{\mu}$  for each lattice structure concisely, we define  $4 \times 4$  matrices as follows

$$\tilde{1}_{\tau\sigma;\tau'\sigma'} \equiv \delta_{\tau\tau'}\delta_{\sigma\sigma'}, \quad (13a)$$

$$\tilde{\boldsymbol{\tau}}_{\tau\sigma;\tau'\sigma'} \equiv \boldsymbol{\sigma}_{\tau\tau'}\delta_{\sigma\sigma'}, \quad (13b)$$

$$\tilde{\boldsymbol{\sigma}}_{\tau\sigma;\tau'\sigma'} \equiv \delta_{\tau\tau'}\boldsymbol{\sigma}_{\sigma\sigma'}, \quad (13c)$$

$$\tilde{\eta}^\pm \equiv (\pm\sqrt{3}\tilde{\tau}^x - \tilde{\tau}^z)/2. \quad (13d)$$

Then, the hopping integrals for the sc lattice are given by

$$t^{(1,0,0)} = [\tilde{1} - \tilde{\eta}^+]t_1, \quad (14a)$$

$$t^{(0,1,0)} = [\tilde{1} - \tilde{\eta}^-]t_1, \quad (14b)$$

$$t^{(0,0,1)} = [\tilde{1} - \tilde{\tau}^z]t_1, \quad (14c)$$

where we have set the lattice constant as unity and  $t_1=3(ff\sigma)/14$ . For the bcc lattice,

$$t^{(1/2,1/2,1/2)} = [\tilde{1} + \tilde{\tau}^y(\tilde{\sigma}^x + \tilde{\sigma}^y + \tilde{\sigma}^z)/\sqrt{3}]t_2, \quad (15a)$$

$$t^{(-1/2,1/2,1/2)} = [\tilde{1} + \tilde{\tau}^y(\tilde{\sigma}^x - \tilde{\sigma}^y - \tilde{\sigma}^z)/\sqrt{3}]t_2, \quad (15b)$$

$$t^{(1/2,-1/2,1/2)} = [\tilde{1} + \tilde{\tau}^y(-\tilde{\sigma}^x + \tilde{\sigma}^y - \tilde{\sigma}^z)/\sqrt{3}]t_2, \quad (15c)$$

$$t^{(1/2,1/2,-1/2)} = [\tilde{1} + \tilde{\tau}^y(-\tilde{\sigma}^x - \tilde{\sigma}^y + \tilde{\sigma}^z)/\sqrt{3}]t_2, \quad (15d)$$

with  $t_2=2(ff\sigma)/21$ . For the fcc lattice,

$$t^{(0,1/2,1/2)} = [\tilde{1} + (\tilde{\eta}^+ - 4\sqrt{3}\tilde{\tau}^y\tilde{\sigma}^x)/7]t_3, \quad (16a)$$

$$t^{(1/2,0,1/2)} = [\tilde{1} + (\tilde{\eta}^- - 4\sqrt{3}\tilde{\tau}^y\tilde{\sigma}^y)/7]t_3, \quad (16b)$$

$$t^{(1/2,1/2,0)} = [\tilde{1} + (\tilde{\tau}^z - 4\sqrt{3}\tilde{\tau}^y\tilde{\sigma}^z)/7]t_3, \quad (16c)$$

$$t^{(0,1/2,-1/2)} = [\tilde{1} + (\tilde{\eta}^+ + 4\sqrt{3}\tilde{\tau}^y\tilde{\sigma}^x)/7]t_3, \quad (16d)$$

$$t^{(-1/2,0,1/2)} = [\tilde{1} + (\tilde{\eta}^- + 4\sqrt{3}\tilde{\tau}^y\tilde{\sigma}^y)/7]t_3, \quad (16e)$$

$$t^{(1/2,-1/2,0)} = [\tilde{1} + (\tilde{\tau}^z + 4\sqrt{3}\tilde{\tau}^y\tilde{\sigma}^z)/7]t_3, \quad (16f)$$

with  $t_3=(ff\sigma)/8$ . Except for the sc lattice, the hopping integrals are complex numbers and dependent on  $\sigma$ . Note that  $t^{-\boldsymbol{\mu}} = t^\boldsymbol{\mu}$ .

### IV. MULTIPOLE INTERACTION

By employing the second-order perturbation theory with respect to  $H_{\text{kin}}$ , we derive the effective Hamiltonian:

$$\begin{aligned} H^{(\text{eff})} &= \sum_{a,b,u} \sum_{m \neq 0} |0,a\rangle\langle 0,a| H_{\text{kin}} \frac{|m,u\rangle\langle m,u|}{E_0 - E_m} \\ &\quad \times H_{\text{kin}} |0,b\rangle\langle 0,b|. \end{aligned} \quad (17)$$

Here,  $|0,a\rangle$  is a ground state without  $H_{\text{kin}}$  with the energy  $E_0$  and  $|m,u\rangle$  is an  $m$ -th excited state with the energy

$E_m$ . In the following, we consider only the first excited states among the intermediate states, which are described by a pair of nearest-neighboring  $f^1$  and  $f^3$  sites discussed above. Then, we need to evaluate the following matrix element:

$$\begin{aligned} & -\Delta E \times H_{\tau_1\tau_2;\tau'_1\tau'_2}^{(\text{eff})}(\mathbf{r}_1, \mathbf{r}_2) \\ &= \sum_u \langle \tau_1(\mathbf{r}_1) \tau_2(\mathbf{r}_2) | H_{\text{kin}} | 1, u \rangle \\ & \quad \times \langle 1, u | H_{\text{kin}} | \tau'_1(\mathbf{r}_1) \tau'_2(\mathbf{r}_2) \rangle, \end{aligned} \quad (18)$$

where  $\Delta E = E_1 - E_0 = J_{78}/2$ . This matrix element denotes the transitions of the states:  $\tau'_1 \rightarrow \tau_1$  at  $\mathbf{r}_1$  and  $\tau'_2 \rightarrow \tau_2$  at  $\mathbf{r}_2$ . The part of the element in which the intermediate  $f^1$  state is located at  $\mathbf{r}_2$  and  $f^3$  state is located at  $\mathbf{r}_1$  is given by

$$\begin{aligned} & \sum_u \langle \tau_1(\mathbf{r}_1) \tau_2(\mathbf{r}_2) | c_{\mathbf{r}_2\nu_2}^\dagger t_{\nu_2\nu_1}^{\mathbf{r}_2-\mathbf{r}_1} c_{\mathbf{r}_1\nu_1} | 1, u \rangle \\ & \quad \times \langle 1, u | c_{\mathbf{r}_1\nu'_1}^\dagger t_{\nu'_1\nu'_2}^{\mathbf{r}_1-\mathbf{r}_2} c_{\mathbf{r}_2\nu'_2} | \tau'_1(\mathbf{r}_1) \tau'_2(\mathbf{r}_2) \rangle \\ &= t_{\nu_2\nu_1}^{\mathbf{r}_2-\mathbf{r}_1} t_{\nu'_1\nu'_2}^{\mathbf{r}_1-\mathbf{r}_2} \\ & \quad \times \langle \tau_1(\mathbf{r}_1) | c_{\mathbf{r}_1\nu_1} | \tilde{\sigma}_1(\mathbf{r}_1) \rangle \langle \tilde{\sigma}_1(\mathbf{r}_1) | c_{\mathbf{r}_1\nu'_1}^\dagger | \tau'_1(\mathbf{r}_1) \rangle \\ & \quad \times \langle \tau_2(\mathbf{r}_2) | c_{\mathbf{r}_2\nu_2}^\dagger | \sigma_2(\mathbf{r}_2) \rangle \langle \sigma_2(\mathbf{r}_2) | c_{\mathbf{r}_2\nu'_2} | \tau'_2(\mathbf{r}_2) \rangle \\ &= t_{\nu_2\nu_1}^{\mathbf{r}_2-\mathbf{r}_1} t_{\nu'_1\nu'_2}^{\mathbf{r}_1-\mathbf{r}_2} \tilde{B}_{\nu'_1\sigma_1}^{\tau_1*} \tilde{B}_{\nu'_2\sigma_2}^{\tau'_1} B_{\nu_2\sigma_2}^{\tau_2*} B_{\nu_1\sigma_1}^{\tau'_2} \\ &= \text{Tr}[B^{\tau_2 T} t^{\mathbf{r}_2-\mathbf{r}_1} \tilde{B}^{\tau_1} \tilde{B}^{\tau'_1 T} t^{\mathbf{r}_1-\mathbf{r}_2} B^{\tau'_2}], \end{aligned} \quad (19)$$

where Tr and T denote trace and transpose of a matrix, respectively. Similarly, the part of the element with the intermediate  $f^1$  state at  $\mathbf{r}_1$  and  $f^3$  state at  $\mathbf{r}_2$  is given by

$$\begin{aligned} & \sum_u \langle \tau_1(\mathbf{r}_1) \tau_2(\mathbf{r}_2) | c_{\mathbf{r}_1\nu'_1}^\dagger t_{\nu'_1\nu'_2}^{\mathbf{r}_1-\mathbf{r}_2} c_{\mathbf{r}_2\nu'_2} | 1, u \rangle \\ & \quad \times \langle 1, u | c_{\mathbf{r}_2\nu_2}^\dagger t_{\nu_2\nu_1}^{\mathbf{r}_2-\mathbf{r}_1} c_{\mathbf{r}_1\nu_1} | \tau'_1(\mathbf{r}_1) \tau'_2(\mathbf{r}_2) \rangle \\ &= t_{\nu_2\nu_1}^{\mathbf{r}_2-\mathbf{r}_1} t_{\nu'_1\nu'_2}^{\mathbf{r}_1-\mathbf{r}_2} \\ & \quad \times \langle \tau_1(\mathbf{r}_1) | c_{\mathbf{r}_1\nu'_1}^\dagger | \sigma_1(\mathbf{r}_1) \rangle \langle \sigma_1(\mathbf{r}_1) | c_{\mathbf{r}_1\nu_1} | \tau'_1(\mathbf{r}_1) \rangle \\ & \quad \times \langle \tau_2(\mathbf{r}_2) | c_{\mathbf{r}_2\nu_2}^\dagger | \tilde{\sigma}_2(\mathbf{r}_2) \rangle \langle \tilde{\sigma}_2(\mathbf{r}_2) | c_{\mathbf{r}_2\nu'_2} | \tau'_2(\mathbf{r}_2) \rangle \\ &= t_{\nu_2\nu_1}^{\mathbf{r}_2-\mathbf{r}_1} t_{\nu'_1\nu'_2}^{\mathbf{r}_1-\mathbf{r}_2} B_{\nu'_1\sigma_1}^{\tau_1*} \tilde{B}_{\nu'_2\sigma_2}^{\tau'_1} \tilde{B}_{\nu_2\sigma_2}^{\tau_2*} \tilde{B}_{\nu_1\sigma_1}^{\tau'_2} \\ &= \text{Tr}[\tilde{B}^{\tau_2 T} t^{\mathbf{r}_2-\mathbf{r}_1} B^{\tau_1} \tilde{B}^{\tau'_1 T} t^{\mathbf{r}_1-\mathbf{r}_2} \tilde{B}^{\tau'_2}] \\ &= \text{Tr}[\tilde{B}^{\tau_2 T} t^{\mathbf{r}_2-\mathbf{r}_1*} B^{\tau_1} B^{\tau'_1 T} t^{\mathbf{r}_1-\mathbf{r}_2*} \tilde{B}^{\tau'_2}]. \end{aligned} \quad (20)$$

Then, the total matrix element of the effective Hamiltonian is

$$\begin{aligned} & -\Delta E \times H_{\tau_1\tau_2;\tau'_1\tau'_2}^{(\text{eff})}(\mathbf{r}_1, \mathbf{r}_2) \\ &= \text{Tr}[B^{\tau_2 T} t^{\mathbf{r}_2-\mathbf{r}_1} \tilde{B}^{\tau_1} \tilde{B}^{\tau'_1 T} t^{\mathbf{r}_1-\mathbf{r}_2} B^{\tau'_2}] \\ & \quad + \text{Tr}[\tilde{B}^{\tau_2 T} t^{\mathbf{r}_2-\mathbf{r}_1*} B^{\tau_1} B^{\tau'_1 T} t^{\mathbf{r}_1-\mathbf{r}_2*} \tilde{B}^{\tau'_2}]. \end{aligned} \quad (21)$$

By straightforward algebraic calculations of the matrices  $B^\tau$ ,  $\tilde{B}^\tau$ , and  $t^\mu$ , we can evaluate this equation for each lattice structure.

The obtained effective Hamiltonian can be rewritten by using the multipole operators for the  $\Gamma_3$  state defined by

$$O_{2\mathbf{r}}^0 = \sum_{\tau\tau'} |\tau(\mathbf{r})\rangle \sigma_{\tau\tau'}^z \langle \tau'(\mathbf{r})|, \quad (22a)$$

$$O_{2\mathbf{r}}^2 = \sum_{\tau\tau'} |\tau(\mathbf{r})\rangle \sigma_{\tau\tau'}^x \langle \tau'(\mathbf{r})|, \quad (22b)$$

$$T_{xy\mathbf{z}\mathbf{r}} = \sum_{\tau\tau'} |\tau(\mathbf{r})\rangle \sigma_{\tau\tau'}^y \langle \tau'(\mathbf{r})|. \quad (22c)$$

$O_{2\mathbf{r}}^0$  and  $O_{2\mathbf{r}}^2$  are the quadrupole moments with  $\Gamma_{3g}$  symmetry and  $T_{xy\mathbf{z}\mathbf{r}}$  is the octupole moment with  $\Gamma_{2u}$  symmetry.

In the following, we show the derived multipole interactions in the Fourier transformed form. Previously, we have also derived the multipole interactions for  $f^1$  systems with the  $\Gamma_8$  CEF ground state by a similar method.<sup>3,4</sup> We will compare the multipole interactions for the present  $f^2$ - $\Gamma_3$  model with those for the  $f^1$ - $\Gamma_8$  model.

#### A. sc lattice

For the sc lattice, we obtain only the following quadrupole interaction,

$$\begin{aligned} H^{(\text{eff})} &= \frac{3}{2} \sum_{\mathbf{q}} \left[ \cos q_z O_{2\mathbf{q}}^0 O_{2-\mathbf{q}}^0 \right. \\ & \quad + \cos q_x \frac{1}{4} (\sqrt{3} O_{2\mathbf{q}}^2 - O_{2\mathbf{q}}^0) (\sqrt{3} O_{2-\mathbf{q}}^2 - O_{2-\mathbf{q}}^0) \\ & \quad \left. + \cos q_y \frac{1}{4} (\sqrt{3} O_{2\mathbf{q}}^2 + O_{2\mathbf{q}}^0) (\sqrt{3} O_{2-\mathbf{q}}^2 + O_{2-\mathbf{q}}^0) \right], \end{aligned} \quad (23)$$

in the unit of  $t_1^2/\Delta E$ . We can intuitively understand why this interaction is dominant since the  $z$  direction is congenial to  $3z^2 - r^2$  ( $O_{2\mathbf{q}}^0$ ) symmetry [see Fig. 2(a)]. Also in the  $\Gamma_8$  model, this quadrupole interaction is the main interaction and the  $\Gamma_{2u}$  octupole interaction is absent.

Note that the derived model is the same as a model for ferromagnetic insulating manganites describing only the orbital degrees of freedom of  $e_g$  electrons,<sup>22-25</sup> except for the overall coefficient. This model has continuously degenerate ground states in the mean-field level due to the frustration which originates from the anisotropic interaction.

If we approximate the ordering vector for  $\text{PrPb}_3$  by  $\mathbf{q} = (\pi, \pi, 0)$  and assume this ordering vector to the model, we obtain an ordering of the  $O_2^2$  moment by the mean-field theory. This is out of accord with the experimental indications of the  $O_2^0$  ordering.<sup>13,26</sup> For the  $O_2^0$  ordering in  $\text{PrPb}_3$ , we need to improve the present theory, for example, by considering the long-range interactions, which are also important to stabilize the incommensurate ordering observed in  $\text{PrPb}_3$ .

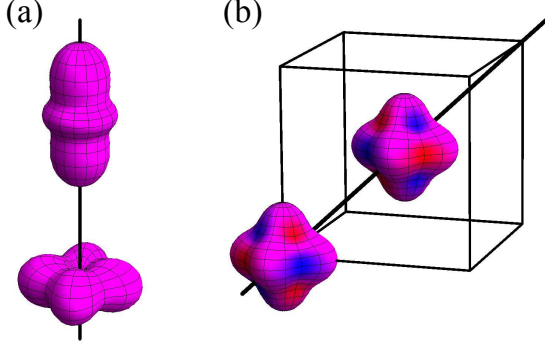


FIG. 2. (Color online) Schematic figures of the electronic states on the nearest-neighboring sites preferred by the interaction (a) along the  $z$  direction (antiferro arrangement of the  $O_2^0$  moments) and (b) along  $[111]$  direction (antiferro arrangement of the  $T_{xyz}$  moments). The gradation of color in (b) indicates the anisotropic distribution of the dipole moment.

### B. bcc lattice

For the bcc lattice, we obtain only the following octupole interaction,

$$H^{(\text{eff})} = 6 \sum_{\mathbf{q}} \cos(q_x/2) \cos(q_y/2) \cos(q_z/2) \times T_{xyz\mathbf{q}} T_{xyz-\mathbf{q}}, \quad (24)$$

in the unit of  $t_2^2/\Delta E$  and the ground state of this effective model is the staggered ordered state of the octupole moments. Since the  $[111]$  direction is congenial to  $xyz$  symmetry, we can naturally understand that this interaction is dominant [see Fig. 2(b)]. Also in the  $\Gamma_8$  model, this octupole interaction is the main interaction and the  $\Gamma_{3g}$  quadrupole interaction is absent. The existence of the  $\tilde{\tau}_y$  term in Eqs. (15a)–(15d) suggests the interaction of the  $T_{xyz}$  moments [Eq. (22c)], in accord with the present result. We will discuss this point in the next subsection.

If ordering of this type of octupole moments occurs, we will observe an anomaly in the specific heat as in an ordinary phase transition, but the determination of the order parameter will be challenging since neither the dipole nor quadrupole moments will be induced, in contrast to the octupole order in  $\text{NpO}_2$  and in  $\text{Ce}_x\text{La}_{1-x}\text{B}_6$  where quadrupole moments are induced.<sup>1–8,12</sup>

The possibility of the ordering of this octupole moment had also been discussed for an  $e_g$ -electron model for manganites in a ferromagnetic metallic phase.<sup>27–30</sup> However, it was revealed that this ordering is unstable against fluctuations beyond the mean-field theory.<sup>31</sup> On the other hand, in the present model for  $f$  electrons, we have a clear picture for the realization of the  $T_{xyz}$  ordering and it should be stable against fluctuations.

Note also that, in the diamond structure, the nearest-neighbor sites locate  $(1/4, 1/4, 1/4)$  and so on from the

origin, and thus, we obtain only the  $\Gamma_{2u}$  octupole interaction as in the bcc lattice. Therefore, we may expect strong fluctuations of the octupole moments in the Pr 1-2-20 systems, in which Pr ions form the diamond structure.

### C. fcc lattice

For the fcc lattice, we obtain both quadrupole and octupole interactions,

$$H^{(\text{eff})} = \frac{3}{49} \sum_{\mathbf{q}} \left[ \cos(q_x/2) \cos(q_y/2) O_{2\mathbf{q}}^0 O_{2-\mathbf{q}}^0 + \cos(q_y/2) \cos(q_z/2) \times \frac{1}{4} (\sqrt{3} O_{2\mathbf{q}}^2 - O_{2\mathbf{q}}^0) (\sqrt{3} O_{2-\mathbf{q}}^2 - O_{2-\mathbf{q}}^0) + \cos(q_z/2) \cos(q_x/2) \times \frac{1}{4} (\sqrt{3} O_{2\mathbf{q}}^2 + O_{2\mathbf{q}}^0) (\sqrt{3} O_{2-\mathbf{q}}^2 + O_{2-\mathbf{q}}^0) \right] \quad (25)$$

$$+ \frac{144}{49} \sum_{\mathbf{q}} [\cos(q_x/2) \cos(q_y/2) + \cos(q_y/2) \cos(q_z/2) + \cos(q_z/2) \cos(q_x/2)] T_{xyz\mathbf{q}} T_{xyz-\mathbf{q}},$$

in the unit of  $t_3^2/\Delta E$ . Broadly speaking, the fcc lattice has characteristics between the sc and bcc lattices and, as a result, we have obtained both quadrupole and octupole interactions. In the  $\Gamma_8$  model, the  $\Gamma_{2u}$  octupole interaction competes with a  $\Gamma_{4u}$  dipole and octupole interaction and a  $\Gamma_{5u}$  octupole interaction, which are absent here since the  $\Gamma_3$  doublet does not have these degrees of freedom. The  $\Gamma_{3g}$  quadrupole interaction is weak but finite in the  $\Gamma_8$  model and it is also similar to the present  $\Gamma_3$  model. Since the octupole interaction is larger than the quadrupole interaction, the ground state of the model is the staggered ordered state of the octupole moments at least in the mean-field theory within two-sublattice structures.

In general, the quadrupole and octupole interactions may compete with each other, but at least in the present simple model, the octupole interaction is dominant. The large difference in the magnitude of the interactions originates from the coefficients in the hopping integral. The ratio of the coefficient of  $\tilde{\eta}^+$  to that of  $\tilde{\tau}^y$  is 1 to  $-4\sqrt{3}$  in Eq. (16a) and the ratio of the square of them is 1 to 48; it is the ratio of the quadrupole and octupole interactions.

In the sc lattice, the hopping integral does not have a  $\tilde{\tau}^y$  term and the octupole interaction is absent. In the bcc lattice, the hopping integral does not have an  $\tilde{\eta}^+$ ,  $\tilde{\eta}^-$  or  $\tilde{\tau}^z$  term and the quadrupole interaction is absent. However, in general, it is not so simple. For example, if the hopping is isotropic, that is, there is no  $\tilde{\eta}^+$ ,  $\tilde{\eta}^-$ ,  $\tilde{\tau}^z$ , or  $\tilde{\tau}^y$  term, we obtain an isotropic Heisenberg-type interaction, i.e., both quadrupole and octupole interactions.

TABLE I. Dominant interactions in each lattice for the  $f^2$ - $\Gamma_3$  model (present study) and for the  $f^1$ - $\Gamma_8$  model (Refs. 3 and 4).

CEF state	sc	bcc	fcc
$f^2$ - $\Gamma_3$	$\Gamma_{3g}$ quadrupole	$\Gamma_{2u}$ octupole	$\Gamma_{2u}$ octupole
$f^1$ - $\Gamma_8$	$\Gamma_{3g}$ quadrupole	$\Gamma_{2u}$ octupole	$\Gamma_{2u}, \Gamma_{4u}, \Gamma_{5u}$

In Table I, we summarize the dominant interactions in each lattice for the  $f^2$ - $\Gamma_3$  model obtained here and for the  $f^1$ - $\Gamma_8$  model (Refs. 3 and 4).

## V. MULTIPOLE INTERACTIONS IN ANOTHER SIMPLIFIED MODEL

In this section, we discuss another simple model to describe the  $\Gamma_3$  CEF ground state. Here, we omit the  $\Gamma_7$  orbital and construct the  $\Gamma_3$  states only from the  $\Gamma_8$  orbitals. This model is too simple to discuss realistic situations, but by comparing with the results in the previous section, we can recognize how much the multipole interactions are altered by the choice of the model. By omitting the  $\Gamma_7$  orbital, the derivation of the multipole interactions becomes rather simple since the intermediate  $f^1$ - $f^3$  states do not split.

For the sc lattice, we obtain no multipole interaction, i.e., the second-order perturbation theory merely gives an energy shift. It means that this model is too simple. For the bcc and fcc lattices, we obtain only the octupole interaction. Thus, the dominance of the octupole interaction in the bcc and fcc lattices is common between the models in this section and in the previous sections.

Therefore, we expect that the characteristic features of the multipole interactions summarized in Table I will not change, even if we use different ways to construct the  $\Gamma_3$  state, except for special cases such as the sc lattice in this section.

## VI. SUMMARY

We have investigated the multipole interactions by the second-order perturbation theory to a simple model for the  $f^2$  ions with the  $\Gamma_3$  non-Kramers doublet ground state under a cubic CEF, in particular, by paying attention to the lattice structure. We have obtained the  $\Gamma_{3g}$  quadrupole interaction for a sc lattice and the  $\Gamma_{2u}$  octupole interaction for a bcc lattice. For an fcc lattice, we have obtained both interactions. These characteristics are the same as those for the  $f^1$ - $\Gamma_8$  model. Thus, we expect that such tendencies or correspondences between the dominant multipole interactions and the lattice structures are common as long as the ground CEF state has these multipole degrees of freedom.

While several kinds of multipole order are possible to occur in general, the  $\Gamma_{2u}$  octupole order is particularly fascinating since it will induce neither the dipole nor quadrupole moments, even though the specific heat will show an anomaly at the transition point as in an ordinary phase transition. In this regard, it would be interesting to search bcc lattices and diamond structure for the  $\Gamma_{2u}$  order since we have obtained a strong interaction for this kind of moments both in the  $f^2$ - $\Gamma_3$  and  $f^1$ - $\Gamma_8$  models.

The general forms of the multipole interactions have been derived in Ref. 32. For example, another form of the quadrupole interaction is possible for a sc lattice in general. We expect that such components appear when we introduce hopping integrals other than  $(ff\sigma)$ . Thus, we should note that the applicability of the present results are limited to the cases where the (effective) hopping processes are mainly described by  $(ff\sigma)$ .

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